# Acoustics of a bubbly fluid at large bubble concentration

Claude Boutin, Jean-Louis Auriault

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ABSTRACT. – The homogenization process is used to investigate how acoustic waves propagate in a bubbly fluid at *finite* concentration. This method involves the considration of waves whose lengths are large compared with the bubble size. We focus on the linear domain, with viscous, thermal and capillary effects. It is shown that this medium displays three different macroscopic descriptions according to bubble diameter and wave frequency. For bubbles of small size, the capillary effect leads to a model where in acoustic waves are either propagative with low celerity or diffusive. On increasing the bubble diameter we obtain waves which are damped and dispersed by both viscous and thermal effects. Finally for large bubbles, we get biphasic undamped and dispersed waves. Moreover, the homogenization method allows us to predict the kind of behaviour and the accuracy of the model from a knowledge of the physical characteristics of the mixture. Applications of these results are presented in the case of water containing air bubbles in equal proportion.

## 1. Introduction

This paper deals with the linear acoustic of bubbly fluids with a finite concentration of bubbles. We define the different macroscopic behaviours of the mixture and we specify their areas of validity in terms of the frequency and the bubble size. The effects of viscosity, thermal conduction and capillarity are all taken into account in the macroscopic description. To keep it simple, the effects of possible mass transfers are not included in this model.

Numerous experimental studies on a liquid-gas mixture under steady of shock waves have been performed [Catersen & Foldy, 1947], [Fox *et al.*, 1955], [Noordjiz & Van Wijingaarden, 1974], [Micaelli, 1982], [Nigmatulin, 1987], [Coste *et al.*, 1990]. Phenomenologic theoretical approaches have been proposed for example in [C & F, 1947], or in [Crespo, 1969] where the effect of the displacement between both phases is discussed,

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and in [Prosperetti, 1976] and [Miksis & Ting, 1984] where thermal effects are taken into account. Currently the homogenization method is leading to new developments in the linear case of a perfect fluid with a finite or infinitesimal gas concentration [Caflish *et al.*, 1985]. In the non-linear area, a model including asymmetric bubble deformations has been proposed by [Miksis & Ting, 1986], and completed in [Miksis & Ting, 1987] by the introduction of a viscous effect in a mixture at weak concentration. The mixtures of two viscous fluids has been treated in [Levy, 1981] and in [Auriault & Lebaigue, 1989] where capillary effects are taken into account. Suspensions of solid particles are studied in [Fleury, 1980].

The homogenization method is suitable for bubbly fluids since they present two well distinct characteristic lengths, the bubble diameter 2R and the macroscopic wave length $\lambda$. In the case of finite concentrations, the *interbubble distance and the size of the bubbles are of the same order of magnitude*, so that the bubble interactions are not overlooked. The influence of viscosity, thermal conduction and capillarity are explitly taken into account. Obviously the physics at the microscopic level is strongly dependent on the physical properties of both elements. Consequently, we have systematically evaluated the differences in the properties of the liquid and the gas by using the ratio $\varepsilon$ between the two characteristic lengths. This procedure allows us to estimate correctly the importance of the different phenomena. In this manner we are able to define correctly the conditions of validity and the accuracy of the macroscopic descriptions.

We describe the principle of the homogenization method in section 2. Section 3 is devoted to the description of the viscous, thermal and capillary effects on the local scale, which allows us to identify the three dimensionless numbers which characterize these phenomena. It appears that these number cannot be varied independently of $\varepsilon$, and that three different cases are possible. Section 4 deals with the results of the homogenization process which is described in the Appendices. It is shown that:

For very small bubbles such as those with $2R \leqq 3\,P^e/\sigma\,\beta$ ($P^e$, $\sigma$, $\beta$ being the equilibrium pressure, the surface tension and the volumic gas concentration respectively), because of the effects of capillarity, the wave celerity decreases with bubble size until it changes from a propagative to a diffusive wave. The gas is under isothermal conditions, and viscosity imposes a bulk displacement of both phases.

When the order of bubble diameter is given by $2R = O(\sqrt{\nu_f/\omega}) \approx O(\sqrt{d_g/\omega})$ ($\nu_f$ being the fluid kinematic viscosity and $d_g$ the thermal diffusivity of the gas), the acoustic waves are dispersive and are damped by viscosity and thermal conduction. Both these effects are of the same order of magnitude, in the case of finite concentration.

When the bubble radius R is of the order of $(\rho_f^e\,\omega^2/12\,\sigma)^{1/3}$ ($\rho_f^e$ being the density of the fluid and $\omega$ the angular frequency of the wave), the gas behaves adiabatically, and the bubbles move in the fluid with asymmetric deformation modes. The coupling between both phases is essentially inertial and it results in undamped but disperive waves.

## 2. Homogenization method

### 2.1. Principle

Let us briefly recall the basic principles of an homogenization process, namely those of multiple scales and asymptotic expansions. For more details one can refer to [Sanchez-Palencia, 1980].

A phenomenon occurring in a thinly heterogeneous medium can be studied using homogenized the equations on condition that there are two very different scales. The first scale characterizes the heterogeneities, and the second is associated with the macroscopic phenomenon. If $l$ and L are the representative lengths of these micro- and macroscopic scales, their ratio $\varepsilon = l/\mathrm{L}$ is a small parameter such as $\varepsilon \ll 1$. Classically, for wave propagation, we have

$$\mathrm{L} = c_0/\omega = \lambda/2\pi$$

where $\lambda$ is the macroscopic wave length and $c_0$ the wave velocity. When taking the microscopic scale to be the diameter of the bubbles ($l \approx 2\mathrm{R}$), we obtain

$$\varepsilon = 2\mathrm{R} \,.\, \omega/c_0.$$

As a consequence of these two separated scales, a spatial invariance is present at the microscopic level. We assume periodic invariance, for two reasons. Firstly, it presents great advantages from a theoretical point of view (the existence and uniqueness of the local fields are guaranteed) and secondly, when the medium is homogenizable, it seems that the *structure* of the constitutive equations is valid for non-periodic materials [Auriault, 1991]. We refer to the periodic cell as simply the cell.

The existence of two very different typical lengths implies that any quantity $\Phi$ depends *a priori* on two dimensionless space variables $\mathrm{X}/l$ and $\mathrm{X}/\mathrm{L}$, or on two physical space variables $y$ and $x$, describing the microscopic and the macroscopic scale, respectively: $\Phi = \Phi(y, x)$; $x = \varepsilon y$. In this way we adopt the microscopic point of view since the leading space variable is the local variable $y$. So the scaling process of the equations will be performed by using $l$ as the characteristic length. The local spatial invariance implies the periodicity of $\Phi$ in the variable $y$.

The use of these two sets of variables introduces powers of $\varepsilon$ in the equations governing the physics at the local scale (through the spatial derivatives $\partial/\partial y$ changed into $\partial/\partial y + \varepsilon \,.\, \partial/\partial x$). For this reason, we naturally look for $\Phi(y, x)$ in the form of an asymptotic expansion in powers of $\varepsilon$, *i.e.*

$$\Phi(y, x) = \sum \varepsilon^n \Phi^n(y, x).$$

Now one substitutes these expansions in to the scaled equations where the dimenionless numbers are scaled with powers of $\varepsilon$, then identifies the terms at each power of $\varepsilon$ and, finally, one solves the corresponding problems.

### 2.2. The parameter ε

A thorough physical analysis on the microscopic scale is essential in order to obtain consistent macroscopic behaviour. Therefore a correct choice of ε is fundamental because ε is used to estimate the relative importance of the different effects acting in the cell.

From a mathematical point of view, ε is an infinitesimal quantity. The macroscopic description is a limit description, infinitely precise, corresponding to infinitely small heterogeneities with regard to the macroscopic length, or inversely, to an infinitely large macroscopic dimension in comparison with the heterogeneities.

From a physical point of view, ε is not an infinitesimal quantity since it corresponds to the finite ratio between the finite micro and macroscopic sizes. As a consequence, the macroscopic description, at the first order of approximation, is a correct representation only to an accuracy of ε. Note that in the cae of wave propagation ε linearly depends on the frequency. In practice, the value of ε is between about $10^{-4}$ to 0.1, but very weak levels are rarely reached (in the case of a representative volume of 1 $mm^3$, $\varepsilon = 10^{-7}$ implies that the macroscopic length is 10 km!)

So, if any dimensionless number or ratio of microscopic effects is about $10^{-4}$, it must be considered $O(1)$ assuming ε infinitely weak, $O(\varepsilon)$ if $\varepsilon \approx 10^{-4}$ or $O(\varepsilon^2)$ when ε is about $10^{-2}$. Of course, these different estimates lead to different macroscopic descriptions. These various scalings express the fact that a weak phenomenon is not negligible with infinite accuracy but must be neglected when the description becomes less accurate.

Later on we will use these two aspects of ε: the mathematical approach for the resolution of the problems and the physical approach for the scaling of the equations and for the interpretation of the homogenized behaviour.

## 3. The physics at microscopic scale

In this section, we examine the physics of bubbly fluids at the microscopic level. We limit ourselves to the study of harmonic excitations, and we only consider small perturbations. Therefore we systematically use the linearized equations in which the convective terms are neglected. The results so obtained are valid to first order when the displacements are small in comparison with the bubble size.

*Notation* For any variable associated with each phase (indexed by α; $\alpha = f$ for the liquid, $\alpha = g$ for the gas) the index $^t$ is related to the sum of the equilibrium value (indexed by $^e$) and the fluctuation value of this quantity. For example, the total density is:

$$\rho_\alpha^t = \rho_\alpha^e + \rho_\alpha e^{i\omega t}$$

### 3.1. The physics of the phenomenon: a simplified approach

The following elementary description is obtained by neglecting the displacements between both phases, the viscosity, the thermal transfers and the capillarity.

3.1.1. *The relative effects of the properties*

Let us study the case of an acoustic wave propagating in an air-water mixture at finite concentration. The values of the density and the incompressibility of both phases are given below ($\gamma = 1.4$ is the specific heat ratio of air).

| $\rho_f^e$ (kg/m$^3$) | $\rho_g^e$ (kg/m$^3$) | $K_f$ (Pa) | $K_g = \gamma . P^e$ (Pa) |
|---|---|---|---|
| $10^3$ | 1.23 | $2.10^9$ | $1.4 . 10^5$ |

If we consider a typical value of $\varepsilon$ of about $10^{-3}$, the ratios of these properties are given by $\rho_g^e/\rho_f^e = O(\varepsilon)$ and $K_g/K_f = O(\varepsilon^{-1})$.

Moreover as we are dealing with finite concentrations $\beta = O(1)$, the typical lengths of the bubbles and the liquid are of the same order of magnitude and the size $l$ of the cell is of the order of the bubble diameter 2R (*Fig.* 1) :

$$l_f = O(l_g) = O(R), \qquad l = O(2R).$$

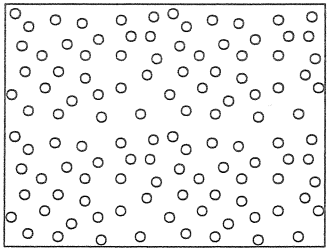

Fig. 1. – A bubbly fluid at finite concentration. The interbubble distance and the bubble radius are of the same order of magnitude.

3.1.2. *Wave propagation*

Under a long wave, the pressure is constant in a cell. Because of the ratio of the compressibility of the phases, the volume variation of the mixture comes from the gas phase only, *i.e.*

$$\Delta V/V = \beta . (\Delta v_g/v_g) = \beta . P/K_g.$$

Therefore the bulk compressibility is equal to $\tilde{K} = K_g/\beta = \gamma . P^e/\beta$, and to the first order of approximation the mixture density is $\tilde{\rho} = (1-\beta)\rho_f^e$.

Consequently the acoustic celerity $c_0$ is approximately given by

$$c_0 = \sqrt{\tilde{K}/\tilde{\rho}} = \sqrt{\gamma . P^e/\beta(1-\beta)\rho_f^e} \text{ and the value of } \varepsilon \text{ is } 2R . \omega/c_0.$$

3.1.3. *Scaling*

From the above simplified analysis, we can define for each quantity its reference value (indexed by $'$) and its dimensionless variable (indexed by $*$), which is necessarily $O(1)$.

*Displacement:* The displacements of both phases are of the same order of magnitude. As we are dealing with small transformations, taking R as the reference length, we have

$$U^r = O(\varepsilon . R), \qquad \text{and} \qquad u_f = U^r . u_f^*, \qquad u_g = U^r . u_g^*.$$

*Densities:* During the propagation of a long wave, the bulk volume variation oscillates on the macroscopic length scale, and the local volume variations are given by

$$(\Delta v_g / v_g) = \varepsilon^{-1} . (\Delta v_f / v_f) = O(U^r/L) = O(\varepsilon . R/L) = O(\varepsilon^2).$$

They are balanced by the density variations, $d(\rho_\alpha^t)/\rho_\alpha^t = O(\Delta v_\alpha / v_\alpha)$.

Therefore, taking $\tilde{\rho}$ as the reference density, we get

$$\rho_f^e = \tilde{\rho} . \rho^{e*},$$

$$\rho_f = \varepsilon^3 . \tilde{\rho} . \rho_f^* \qquad \text{and} \qquad \rho_g^e = \varepsilon . \tilde{\rho} . \rho_g^{e*}, \quad \rho_g = \varepsilon^3 . \tilde{\rho} \rho_g^*.$$

Consequently, the continuity equations are written for both phases in the following linearized and non dimensional forms:

- In the liquid: $\varepsilon^2 (\rho_f^*) + (\rho_f^{e*})\ \mathrm{div}^* (\mathbf{u}_f^*) = 0.$
- In the gas: $\varepsilon (\rho_g^*) + (\rho_g^{e*})\ \mathrm{div}^* (\mathbf{u}_g^*) = 0.$

*Pressures*: The pressure in both phases are of the order of the reference pressure.

$$P^r = \tilde{K} . U^r/L, \qquad P_f = P^r . P_f^*, \qquad P_g = P^r . P_g^*.$$

*Wave propagation:* The phenomenon is propagative on the macroscopic scale so the pressure gradient is balanced by the inertial terms. Therefore we have

$$\mathrm{grad}\ (P) = O(P^r/L) = O(\tilde{\rho} . \omega^2 . U^r).$$

In the next sections we introduce the viscous, thermal and capillarity effects, while keeping the above scalings, which describe the acoustic properties of the mixture.

### 3.2. Viscous effects

Until now we have taken into account neither the relative displacements between the fluid and the gas nor their viscosities. This point has already been studied in [M & T, 1987] in the case of low concentration and in [L, 1981] and [A & L, 1989] for a mixture of two liquids.

#### 3.2.1. *The Navier-Stokes equation*

Under harmonic excitation, and in the linearized form, the Navier-Stokes eqaution becomes

$$\mathbf{grad}\ (P_\alpha) - i\omega\mu_\alpha (\mathbf{grad}\ (\mathbf{div}\ (\mathbf{u}_\alpha)) + \Delta(\mathbf{u}_\alpha)) = \rho_\alpha^t . \omega^2 . \mathbf{u}_\alpha,$$

where $\mu$ is the dynamic viscosity and $\mathbf{u}$ the displacement field.

We see from the table below, that in the case of an air-water mixture, the kinematic viscosities ($\nu = \mu/\rho^e$) of the gas and the liquid are roughly of the same order.

| $\mu_f$ (Pa.s) | $\mu_g$ (Pa.s) | $\nu_f$ ($m^2/s$) | $\nu_g$ ($m^2/s$) |
|---|---|---|---|
| $10^{-3}$ | $19.10^{-6}$ | $10^{-6}$ | $15.10^{-6}$ |

3.2.2. *The Scalings*

We scale the equations by adopting R as the reference length ($y = R.y^*$).

Beginning with the momentum balance *in the fluid*, we divide the Navier-Stokes equation by the reference pressure gradient $P^r/L$ to get

$$\varepsilon^{-1}\,\mathbf{grad}^*(P_f^*) - i.N_v.S.\rho_f^{e*}(\Delta^*(\mathbf{u}_f^*) + \mathbf{grad}^*(\mathrm{div}^*(\mathbf{u}_f^*))) = S.(\rho_f^{e*} + \varepsilon^3.\rho_f^* e^{i\omega t}).\mathbf{u}_f^*,$$

with

$$N_v = \nu_f/\omega R^2,\ S = \tilde{\rho}.\omega^2.U^r/(P^r/L).$$

*In the gas*, the same procedure leads to

$$\varepsilon^{-1}\,\mathbf{grad}^*(P_g^*) - i\varepsilon N_{vg} S.\rho_g^{e*}[\Delta^*(\mathbf{u}_g^*) + \mathbf{grad}^*(\mathbf{div}^*(\mathbf{u}_g^*))] = \varepsilon S(\rho_g^{e*} + \varepsilon^2.\rho_g^* e^{i\omega t})\,\mathbf{u}_g^*,$$

with $N_{vg} = \nu_g/\omega R^2$.

The fluctuations of the stresses in each fluid are given by

$$\sigma_\alpha = -P_\alpha \mathbf{I} + 2i\omega\mu_\alpha \mathbf{E}(\mathbf{u}_\alpha), \qquad \text{with} \quad \mathbf{E}(\mathbf{u})_{ij} = (\mathbf{u}_{i,j} + \mathbf{u}_{j,i})/2.$$

When we normalize these stresses using the reference pressure, we get

$$\sigma_f = P^r.(-P_f^*.\mathbf{I} + 2.\varepsilon.i.N_v.S.\mathbf{E}^*(\mathbf{u}_f^*)) \text{ in the fluid,}$$

and

$$\sigma_g = P^r.(-P_g^*.\mathbf{I} + 2.\varepsilon^2.i(N_{vg}.S)\rho_g^{e*}.\mathbf{E}^*(\mathbf{u}_g^*)) \text{ in the gas.}$$

We have seen in the previous section that the reference pressure gradient is balanced by the inertial forces. Therefore $S = O(1)$.

The ratios between viscous and inertial terms, $N_v$ and $N_{vg}$, can be expressed as

$$N_v = (l_v/R)^2 \quad \text{and} \quad N_{vg} = (l_{vg}/R)^2 \quad \text{with } l_v = \sqrt{\nu_f/\omega} \quad \text{and} \quad l_{vg} = \sqrt{\nu_g/\omega},$$

where $l_v$ and $l_{vg}$ represent the thickness of the viscous layer in the fluid and in the gas respectively. As they are of the same order of magnitude, we will only use $N_v$ to characterize the viscous effects. Their influence is described by the order of magnitude of the dimensionless number $N_V$. In section 4 we study the three more representative cases: $N_V = O(\varepsilon^{-1})$, $N_V = O(1)$, $N_V = O(\varepsilon)$.

In Section 3.1 we used the adiabatical compressibility of the gas. As shown by [P, 1977] or [M & T, 1984], this assumption is not valid in the presence of heat transfer. These effects are described by the Fourier conduction law.

### 3.3.1. *The Fourier law and state equation*

The linearized form of the Fourier law under harmonic excitation is

$$\operatorname{div}(k_\alpha . \mathbf{grad}(T_\alpha)) = i\omega . (\rho^t_\alpha . C_{p\alpha} - P_\alpha),$$

where $k_\alpha$, $C_{p\alpha}$ are the conductivity and the specific heat, respectively.

The thermal constants of air and water are given in the table below. We notice that the conductivities are roughly of the same order, but the fluid diffusivity $d_f = k_f/\rho^e_f . C_{pf}$ is much larger than that of the gas.

| $C_{pf}$ | $C_{pg}$ | $k_f$ | $k_g$ | $d_f$ | $d_g$ |
|---|---|---|---|---|---|
| (J/°K.kg) | (J/°K.kg) | (W/°K.m) | (W/°K.m) | ($m^2$/s) | ($m^2$/s) |
| $4.18.10^3$ | $10^3$ | 0.602 | 0.026 | $2.12.10^{-5}$ | $1.4.10^{-7}$ |

The thickness of the thermal layer is given by $l_{t\alpha} = \sqrt{d_\alpha/\omega}$.

– If this *layer is small* relative to R, the heat transfers are negligible outside this zone, and the transformation is adiabatic. We have $T_\alpha = O(P_\alpha/\rho^t_\alpha . C_{p\alpha})$.

The difference in the two densities means that temperature fluctuations in the gas are much more important than those in the fluid so that $T_g = O(\varepsilon^{-1} . T_f)$.

– on the other hand if the frequency is sufficiently low $l_\alpha$ *is large* in comparison with R, the perturbation occurs isothermally. The continuity of thermal flux allows us to compare the temperature levels in both phases and give $T_g = O(T_f)$.

*Equation of state:* Under small harmonic perturbations, assuming that the gas is perfect, we have

$$P_g = P^e(\rho_g/\rho^e_g + T_g/T^e).$$

### 3.3.2. *The Scalings*

We take as the reference temperature variation $T^r = P^r/\rho^e_g . C_{pg}$.

Under adiabatic conditions we have $T_g = O(T^r)$.

Let us first scale the Fourier equation *in the gas*. Dividing the equation by the thermal inertial terms $\rho^e_g . C_{pg} . \omega . T^r$, we get

$$N_t . \operatorname{div}(k^*_g . \mathbf{grad}(T^*_g)) = i . (T^*_g - P^*_g), \qquad \text{where} \quad N_t = d_g/\omega . R^2 = (l_{tg}/R)^2.$$

Therefore the above dimensionless form of the Fourier equation is scaled for $N_t \leqq O(1)$.

Conversely, when $N_t = O(\varepsilon^{-1})$ and the conditions are isothermal, then $T_g = O(\varepsilon . T^r)$, and we have

$$\varepsilon . N_t . \operatorname{div}(k^*_g . \mathbf{grad}(T^*_g)) = i . (\varepsilon . T^*_g - P^*_g).$$

Considering now *the fluid*, its temperature is of the order of

$$T_f = O((\rho_g^e C_{pg}/\rho_f^e C_{pf})\, T^r) = O(\varepsilon . T^r).$$

If we divide the Fourier equation by the thermal inertial term $\rho_f^e . C_{pf} . \omega . T^r$, we get

$$\varepsilon . N_{tf} . \operatorname{div}(k_f^* . \mathbf{grad}(T_f^*)) = i . \varepsilon . (T_f^* - P_f^*), \qquad \text{where} \quad N_{tf} = d_f/\omega . R^2 = (l_{tf}/R)^2.$$

Since the density is much larger in the fluid we have $N_{tf}/N_t = O(\varepsilon)$.

Hereinafter we use $N_t$ to characterize the thermal effects, and we study the three following cases $N_t = O(\varepsilon^{-1})$, $N_t = O(1)$, $N_t = O(\varepsilon)$.

*The equation of State:* Taking into account the normalisations of the pressures, densities and temperatures, the dimensionless state equation of the gas is written in the form: when $N_t \leqq O(1)$

$$\gamma P_g^* = \rho_f^*/\beta . \rho_g^{e*} + (\gamma - 1)(\rho_g^{e*} C_{pg}/\rho_f^{e*} C_{pf}) T_f^*,$$

and for $N_t \geqq O(\varepsilon)$

$$\gamma P_g^* = \rho_f^*/\beta . \rho_g^{e*} + \varepsilon(\gamma - 1)(\rho_g^{e*} C_{pg}/\rho_f^{e*} C_{pf}) T_f^*.$$

### 3.4. Capillarity effects

We focus here on the effects of the capillarity. If the capillary tension is $\sigma$ (for water $\sigma = 75 . 10^{-3}$ N/m), and the variation of the bubble radius under small perturbations is $r$, then we have on $\Gamma$

$$(\sigma_g - \sigma_f) . \mathbf{n} = P_c . \mathbf{n} \qquad \text{with} \quad P_c = -2\sigma d(1/R) = 2\sigma r/R^2.$$

#### 3.4.1.* *Scaling*

The dimensionless form of this equation is $(\sigma_g^* - \sigma_f^*) . \mathbf{n} = (P_c/P^r) . \mathbf{n}$ with

$$W^{-1} = P_c/P^r = (2\sigma . \beta/3R . K_g)(3 . r/R)(L/U^r).$$

This expression shows that the Weber number $W = P^r/P_c$ depends on two factors.

On the one hand, it depends on the ratio between the *"rigidity"* of the capillary membrane $K_c = 2\sigma/3R$ and the bulk rigidity $K_g/\beta$ (here we take $K_g = P^e$, which we justify later). Again we will have to consider three sizes of the ratio to describe most of the possible cases $K_c . \beta/P^e = O(1)$, $O(\varepsilon)$, $O(\varepsilon^2)$.

On the other hand, W depends on $r/R$, *i.e.* on the deformation mode of the bubbles. Two cases can be considered:

– either the curvature variation is one order of magnitude less than the displacement *i.e.* $r/R = O(\varepsilon^2)$

– or, as proposed in [M & T, 1986], the variation of the curvature radius is of the same order as the displacement, which gives $r/R = O(U^r/R) = O(\varepsilon)$.

As the volume variation is $O(\varepsilon^2)$, the deformation mode must necessarily be asymmetric.

### 3.5. Summary

The microscopic description given in the previous paragraphs is valid for the following conditions concerning the properties of the constituents:

$$\rho_g^e/\rho_f^e = O(\varepsilon); \qquad K_g/K_f = O(\varepsilon); \qquad \nu_g/\nu_f = O(1);$$

$$C_{pg}/C_{pf} = O(1); \qquad k_g/k_f = O(1)$$

The effects of the viscosity, the thermal conduction and the capillary tension, characterized by the dimensionless numbers $N_v$, $N_t$, W respectively, depend not only on the physical constants but also on the bubble size and the excitation frequency.

It is important to notice that the orders of magnitude of these numbers are not independent. As a matter of fact, for air-water mixtures, the thermal layer $l_{tg}$ in the gas and the viscous layer in the liquid $l_{vf}$ are of about the same order of magnitude, because

$$l_t/l_v = \sqrt{d_g/\nu_f} \approx 0.4 = O(1) \quad i.e.\ N_t = O(N_v).$$

Moreover, decreasing or increasing the diameter of the bubbles and their relative separation makes viscous, thermal and capillary terms increase or decrease, respectively. At atmospheric pressure, with the usual values of the physical constants we have

$$c_0 \approx 25 \text{ m/s} \quad \text{so} \quad \sigma/3\mu_f . c_0 = O(1) \qquad \text{and} \qquad \sigma C_{pg}/3 k_g . c_0 = O(1)$$

which leads to the following relationship $K_c . \beta/P^e = O(\varepsilon N_v) = O(\varepsilon N_t)$.

In next section we treat three cases (A, B, C) which cover a large range of bubble diameters ($10^{-1}$ to $10^{-8}$ m) and of acoustic frequencies ($10^{-1}$ to $10^6$ Hz). Table presents for each case the order of magnitude of the dimensionless numbers and gives, in terms of bubbles' size and frequency, the condition of validity for each case.

Table I. – *Summary of the three cases and their conditions of validity.*

| | $N_v$ | $N_t$ | $K_c . \beta/P^e$ |
|---|---|---|---|
| Case B | $O(\varepsilon^{-1})$<br>$\nu_f/\omega . R^2 \approx c_0/2\ R . \omega$ | $O(\varepsilon^{-1})$<br>$d_g/\omega\ R^2 \approx c_0/2\ R . \omega$ | $O(1)$<br>$2\sigma . \beta/3\ R . P^e \approx 1$ |
| Case A | $O(1)$<br>$\nu_f/\omega . R^2 \approx 1$ | $O(1)$<br>$d_g/\omega\ R^2 \approx 1$ | $O(\varepsilon)$<br>$2\sigma . \beta/3\ R . P^e \approx 2\ R . \omega/c_0$ |
| Case C | $O(\varepsilon)$<br>$\nu_f/\omega . R^2 \approx 2\ R . \omega/c_0$ | $O(\varepsilon)$<br>$d_g/\omega\ R^2 \approx 2\ R . \omega/c_0$ | $O(\varepsilon^2)$<br>$2\sigma\ \beta/3\ RP^e \approx (2\ R\ \omega/c_0)^2$ |

## 4. Macroscopic descriptions

We examine here three different macroscopic behaviours of a bubbly fuid:

case A: "Medium" bubble size and frequencies;

case B: "Small" bubble size;

case C: "Large" bubble size and "high" frequencies.

### 4.1. Case A: "Medium" bubble size and frequencies

The case where $N_v = O(N_t) = O(1)$, $K_c . \beta / P^e = O(\varepsilon)$ corresponds to bubbles of a diameter of the order of magnitude of the viscous and the thermal layers, and for which the "rigidity" of the capillary membrane is weak.

#### 4.1.1. *Formal scaled equations*

In order to simplify the presentation we will use the dimensional variables with the formal scaled equations. From the dimensional analysis of Section 3, the system to be treated is

*In the liquid*

$$P_f = -\varepsilon^{-2} . K_f . \operatorname{div}(\mathbf{u}_f), \tag{A.1}$$

$$\sigma_f = -P_f . \mathbf{I} + 2 . i . \varepsilon . \omega . \mu_f . \mathbf{E}(\mathbf{u}_f), \tag{A.2}$$

$$\varepsilon^{-1} . \mathbf{grad}(P_f) - i . \omega . \mu_f . (\Delta(\mathbf{u}_f) + \mathbf{grad}(\operatorname{div}(\mathbf{u}_f))) = \rho_f^e . \omega^2 \mathbf{u}_f, \tag{A.3}$$

$$\varepsilon^2 . \rho_f + \rho_f^e . \operatorname{div}(\mathbf{u}_f) = 0, \tag{A.4}$$

$$\varepsilon . \operatorname{div}(k_f . \mathbf{grad}(T_f)) = i\omega . (\rho_f^e . C_{pf} . T_f - P_f). \tag{A.5}$$

*In the gas*

$$P_g = P^e . (\rho_g / \rho_g^e + T_g / T^e), \tag{A.6}$$

$$\sigma_g = -P_g . \mathbf{I} + 2 . i . \varepsilon^2 . \omega . \mu_g \mathbf{E}(\mathbf{u}_g), \tag{A.7}$$

$$\varepsilon^{-1} . \mathbf{grad}(P_g) - i . \varepsilon . \omega . \mu_g . (\Delta(\mathbf{u}_g) + \mathbf{grad}(\operatorname{div}(\mathbf{u}_f))) = \varepsilon . \rho_g^e . \omega^2 . \mathbf{u}_g, \tag{A.8}$$

$$\varepsilon . \rho_g + \rho_g^e . \operatorname{div}(\mathbf{u}_g) = 0, \tag{A.9}$$

$$\operatorname{div}(k_g . \mathbf{grad}(T_g)) = i\omega . (\rho_g^e . C_{pg} . T_g - P_g). \tag{A.10}$$

*At the interfaces*

$$\mathbf{u}_f = \mathbf{u}_g, \tag{A.11}$$

$$(\sigma_g - \sigma_f) . \mathbf{n} = \varepsilon . P_c . \mathbf{n} \quad \text{with} \quad P_c = 2\sigma r / R^2, \tag{A.12}$$

$$T_f = \varepsilon^{-1} . T_g, \tag{A.13}$$

$$k_f . \mathbf{grad}(T_f) . \mathbf{n} = \varepsilon^{-1} . k_g . \mathbf{grad}(T_g) . \mathbf{n}. \tag{A.14}$$

#### 4.1.2. *Macroscopic behaviour*

The details of the homogenization procedure are given in Appendix A.

As the thickness of viscous layer is of the order of magnitude of the interbubble distance, the velocity field is inhomogeneous within the cell. Consequently, the bubbles move in the fluid and this relative displacement contains a phase shift with respect to the pressure gradient because of the viscosity. Since the diffusivity of the fluid is weak; its temperature is constant in the cell, except in the vicinity of the bubbles. Conversely, the gas temperature is not constant, and the heat flux introduces a phase shift between the pressure and the temperature perturbations. Therefore the volume variations of the gas are not in phase with the pressure. Lastly, the capillary tension constrains the bubble

deformations to be spherical, while being negligible in comparison with the pressure fluctuations.

We show, in Appendix A, that the mixture behaves macroscopically like a medium with one pressure field P and two displacement fields $\mathbf{U}_f$ and $\mathbf{U}_g$, which satisfy:

$$\text{(A.15)} \qquad \begin{cases} (1-\beta).\mathbf{U}_f = (1/\rho_f^e.\omega^2)\,\mathbf{grad}\,(P), \\ \beta.\mathbf{U}_g = (\beta/\rho_f^e.\omega^2)\,(\mathbf{I}+\mathbf{M})\,\mathbf{grad}\,(P), \\ \operatorname{div}((1-\beta).\mathbf{U}_f)+\operatorname{div}(\beta.\mathbf{U}_g) = \beta(-1+G(1-1/\gamma))\,P/P^e. \end{cases}$$

$\mathbf{M}$ is a complex symmetric tensor which depends on the dimensionless frequency $\omega/\omega_v$, $(\omega_v = \nu_f/R^2)$. G is a complex function depending on $\omega/\omega_t$, $(\omega_t = d_g/R^2)$.

*Comments*

– The meaning of the first two equations is clearer if they are written in their inverse forms:

$$(1-\beta).\mathbf{grad}\,(P) = (1-\beta).\rho_f^e.\omega^2\,[(\mathbf{M}.(1-\beta)/\beta-\mathbf{I})^{-1}(\mathbf{U}_f-\mathbf{U}_g)+\mathbf{U}_f]$$

$$\beta.\mathbf{grad}\,(P) = (1-\beta).\rho_f^e.\omega^2\,[-(\mathbf{M}.(1-\beta)/\beta-\mathbf{I})^{-1}(\mathbf{U}_f-\mathbf{U}_g)].$$

The real part of the complex tensor $(\mathbf{M}.(1-\beta)/\beta-\mathbf{I})^{-1}$ is related to the inertial coupling, and its the imaginary part, divided by the frequency corresponds to the viscous coupling. The calculation of tensor $\mathbf{M}$ uses the variational formulation given in Appendix A.

– The complex function G describes the fact that the gas is not in adiabatic or isothermal conditions. This function can be expressed analytically in the form [Auriault, 1983]:

$$G = 1-(3/i\omega_t^*)\,(1+\coth(\sqrt{i\omega_t^*}).\sqrt{i\omega_t^*}) \qquad \text{with} \quad \omega_t^* = \omega/\omega_t.$$

– The medium is dissipative. In the case of *large concentration* the dissipation due to the viscous effects is of the *same order* of magnitude as that due to thermal effects.

– For transient vibrations, this frequency description can be transformed to the temporal domain by an inverse Fourier transform. The frequency dependence of the description introduces memory effects with a duration of about $\tau = O(2\pi/\omega_v) \approx O(2\pi/\omega_t)$.

4.1.3. *Wave propagation*

We focus on harmonic pressure waves, in the case of isotropic macroscopic behaviour $\mathbf{M} = M.\mathbf{I}$. The elimination of $\mathbf{U}_f$ and $\mathbf{U}_g$ in (A.15) leads to the equation

$$\text{(A.16)} \qquad -(1/\rho_f^e.\omega^2)\,(1+\beta(1+M))\,\Delta(P) = \beta(1-G(1-1/\gamma))\,P/P^e.$$

We look for solutions of the form

$$P = P_0.e^{i\omega(t-x/c)}; \qquad c = c_1 + i.c_2.$$

On substituting this expression into (A.16), we get the dispersion equation

$$c^2=(c_1+i.c_2)^2=(c_0)^2\,[1+\beta^2\,(M\,(1-\beta)/\beta-1)]/[\gamma-G\,(\gamma-1)].$$

The present medium produces a dispersive attenuated propagation mode, since M and G are complex and depend on $\omega$. As $M=O(1)$ and $G=O(1)$, the celerity $c$ is of the same order as $c_0$. This result extends that obtained in [P, 1977], [M & T, 1984 (who considered only the thermal effects) and in [M & T, 1987] where only the viscous effects are considered and in the case of weak concentrations so that the bubble interactions are negligible.

### 4.2. Case B: "Small" bubble size

In this case $N_v=O(N_t)=O(\varepsilon^{-1})$ and $K_c.\beta/P^e=O(1)$. The bubble diameter is much smaller than the thicknesses of the viscous and thermal layers, and the "rigidity" of the capillary membrane is of the same order of magnitude as that of the mixture.

#### 4.2.1. *Formal scaled equations*

The set of formal scaled equations to be treated is (A.1-A.14) with the exception of the following which are now indexed with B.

*In the liquid*

$$\varepsilon^{-1}\,(\mathbf{grad}\,(P_f)-i.\varepsilon^{-1}.\omega.\mu_f.(\Delta\,(\mathbf{u}_f)+\mathbf{grad}\,(\mathrm{div}\,(\mathbf{u}_f)))=\rho_f^e.\omega^2.\mathbf{u}_f, \tag{B.3}$$

$$\mathrm{div}\,(k_f.\mathbf{grad}\,(T_f))=i\omega.(\rho_f^e.C_{pf}.T_f-P_f). \tag{B.5}$$

*In the gas*

$$P_g=P^e.(\rho_g/\rho_g^e+\varepsilon.T_g/T^e), \tag{B.6}$$

$$\sigma_g=-P_g.\mathbf{I}+2.i.\varepsilon.\omega.\mu_g\,\mathbf{E}\,(\mathbf{u}_g), \tag{B.7}$$

$$\varepsilon^{-1}.\mathbf{grad}\,(P_g)-i.\omega.\mu_g.(\Delta\,(\mathbf{u}_g)+\mathbf{grad}\,(div\,(\mathbf{u}_f)))=\varepsilon.\rho_g^e.\omega^2.\mathbf{u}_g, \tag{B.8}$$

$$\varepsilon^{-1}.\mathrm{div}\,(k_g.\mathbf{grad}\,(T_g))=i\omega.(\rho_g^e.C_{pg}.T_g-P_g). \tag{B.10}$$

*At the interfaces*

$$(\sigma_g-\sigma_f).\mathbf{n}=P_c.\mathbf{n}, \tag{B.12}$$

$$T_f=T_g, \tag{B.13}$$

$$k_f.\mathbf{grad}\,(T_f).\mathbf{n}=k_g.\mathbf{grad}\,(T_g).\mathbf{n}. \tag{B.14}$$

#### 4.2.2. *Macroscopic description*

As the viscous layer is much larger than the interbubble distance, the bubbles and the fluid more together, in phase with the pressure gradient. Moreover, the bubble size is very small in comparison with the thermal layer. Therefore the temperature is constant in the gas, which corresponds to isothermal conditions. Lastly the capillary tension imposes spherical deformations, and is of the same order of magnitude as the pressure spherical deformations, and is of the same order of magnitude as the pressure perturbations. Therefore the pressure in each phase is different.

The homogenization (Appendix B) shows that the mixture behaves likes a medium with one displacement field U and two pressure fields $P_f$ and $P_g$ satisfying the equations:

$$(B.15)\quad \begin{cases} P_g = -(P^e/\beta)\,\mathrm{div}\,(U), \\ P_f = -(1-(1-\beta).K_c/P^e)(P^e/\beta)\,\mathrm{div}\,(U), \qquad \text{with} \quad K_c = 2\sigma/3R, \\ \mathbf{grad}\,(P_f) + \beta/(1-\beta)\,\mathbf{grad}\,(P_g) = (\rho_f^e.\omega^2)\,U. \end{cases}$$

*Comments*

– At the first order of approximation, the medium is not dissipative. This comes from the isothermal conditions of the gas and from the viscosity which stops the relative displacements whithin the cell. (As a matter of fact, the medium is dissipative, but at second order only)

– The equivalent medium is isotropic whatever the arrangement of the bubbles in the cell.

– The description does not depend on the frequency, so it remains valid for transient motions.

4.2.3. *Wave propagation*

Let us study harmonic wave propagation. Looking for a solution for the gas pressure in the form

$$P_g = P_0.e^{i\omega(t-x/c)},$$

Eqs. (B.15 *a* and *b*) give $P_f = (1-(1-\beta).K_c/P^e)P_g$.

Then, eliminating the displacement U in (B.15 *c*) gives

$$(1-\beta).\Delta(P_f) + \beta.\Delta(P_g) = -((1-\beta).\rho_f^e.\omega^2)(\beta/P^e)P_g,$$

leading to

$$c^2 = (1-(1-\beta)^2.(K_c/P^e))(P^e/(1-\beta).\beta.\rho_f^e) = [(1-(1-\beta)^2.(K_c/P^e))/\gamma]\,c_0^2.$$

There we have to distinguish between different kinds of propagation according to the value of $K_c$, which is directly related to the bubbles radius since $K_c = 2\sigma/3R$.

– If $K_c < P^e/(1-\beta)$, we obtain a real celerity, independent of the frequency

$$c = c_0.[(1-(1-\beta)^2.(K_c/P^e))/\gamma]^{1/2}.$$

The celerity is smaller than $c_0$ because of the capillarity and the isothermal conditions in the gas. As the wave travels through the medium, the pressure in the fluid and in the gas are in phase, but the capillary tension imposes less pressure in the fluid.

– If $P^e/(1-\beta) \leqq K_c \leqq P^e/(1-\beta)^2$, then the velocity becomes very small (theoretically zero when $K_c = P^e/(1-\beta)^2$). The capillary tension, which almost compensates for the gas pressure, prevents the propagation of the acoustic wave. Moreover, due to the small value of the celerity, diffraction effects due to the bubbles can occur.

– If $K_c > P^2/(1-\beta)^2$, $c^2$ is a *negative number*, the celerity is purely imaginary

$$c = i.c_0.\left|(1-(1-\beta)^2.(K_c/P^e))/\gamma\right|^{1/2}.$$

Therefore the wave is no longer propagative but *diffusive*. The propagation is prevented by the "negative rigidity" of the capillary membrane which causes the gas and liquid pressures to be opposite in phase.

In short, we will bear in mind that for "little" bubbles, , *i.e.* if $2R \approx 4\sigma(1-\beta)^2/3P^e$, the capillarity is an essential factor for the propagation of acoustic waves in bubbly fluids.

### 4.3. Case C: "Large" bubble size and "high" frequencies

Assuming bubbles of "large" radii, the capillary and viscous effects become negligible to first order. Therefore we have $N_v = O(N_t) = O(\varepsilon)$ and $K_c.\beta/P^e = O(\varepsilon^2)$.

Due to the weak rigidity of the capillary membrane, the deformation mode of the bubbles can be assymmetrical. In such conditions, the bubble curvature variation, $r$, is of the same order as the displacement, so $r/R = O(\varepsilon)$ and $W = O(\varepsilon)$.

#### 4.3.1. *Formal scaled equations*

The relevant equations are (A.1-A.14) except for the equations below indexed with C.

*In the liquid*

(C.2) $$\sigma_f = -P_f.\mathbf{I} + 2.i.\varepsilon^2.\omega.\mu_f.\mathbf{E}(\mathbf{u}_f),$$

(C.3) $$\varepsilon^{-1}.\mathbf{grad}(P_f) - i.\omega.\varepsilon.\mu_f.(\Delta(\mathbf{u}_f) + \mathbf{grad}(\operatorname{div}(\mathbf{u}_f))) = \rho_f^e.\omega^2.\mathbf{u}_f,$$

(C.5) $$\varepsilon^2.\operatorname{div}(k_f.\mathbf{grad}(T_f)) = i\omega.(\rho_f^e.C_{pf}.T_f - P_f).$$

*In the gas*

(C.7) $$\sigma_g = -P_g.\mathbf{I} + 2.i.\varepsilon^2.\omega.\mu_g\,\mathbf{E}(\mathbf{u}_g),$$

(C.8) $$\varepsilon^{-1}.\mathbf{grad}(P_g) - i.\omega.\mu_g.\varepsilon^2(\Delta(\mathbf{u}_g) + \mathbf{grad}(\operatorname{div}(\mathbf{u}_f))) = \varepsilon.\rho_g^e.\omega^2.\mathbf{u}_g,$$

(C.10) $$\varepsilon.\operatorname{div}(k_g.\mathbf{grad}(T_g)) = i\omega.(\rho_g^e.C_{pg}.T_g - P_g).$$

#### 4.3.2. *Macroscopic description and wave propagation*

The displacement fields of both phases are constant except within the thin viscous layer located at the boundaries of the bubbles. Consequently, the bubbles move relative to the liquid. This introduces an inertial coupling between the fluid and the gas corresponding to a virtual mass effect. In the same manner the thermal conduction is limited to the vicinity of the bubbles, so the gas is under adiabatic conditions. Lastly the capillary tension is weak enough to allow assymmetrical deformation modes.

By the homogenization process (Appendix C) the medium is described by a single pressure field P and two displacement fields $\mathbf{U}_f$ and $\mathbf{U}_g$, described by the equations below.

(C15)
$$\begin{cases} (1-\beta).\mathbf{U}_f = (1/\rho_f^e.\omega^2)\,\mathbf{grad}(P), \\ \beta.\mathbf{U}_g = (\beta/\rho_f^e.\omega^2)(\mathbf{I}+\mathbf{M}')\,\mathbf{grad}(P), \\ \operatorname{div}((1-\beta).\mathbf{U}_f) + \operatorname{div}(\beta.\mathbf{U}_g) = -(\beta/\gamma P^e)P. \end{cases}$$

*Comments*

– In contrast to the tensor $\mathbf{M}$ in (A.15), the tensor $\mathbf{M}'$ is real Moreover there is no phase shift due to the thermal effect since at first order the gas is under adiabatic conditions.

– The symmetric tensor $\mathbf{M}'$ includes the inertial and capillary effects and depends on the frequency through $(1/2)(\omega_r/\omega)^2$ where $\omega_r = \sqrt{12 . \sigma/\rho_f^e . R^3}$.

$\omega_r$ is the second oscillation frequency of a bubble in a fluid (Lamb cited in [M & T, 1986]).

– Although the bubbles are deformed and move within the cell, the viscous effects are too weak to appear at the macroscopic level. At the first order of approximation, the propagation arises without any attenuation. (The dissipation is of the second order, which introduces an attenuation per wave length of the order of $\varepsilon$, in the real medium).

– In the isotropic case ($\mathbf{M}' = M' . \mathbf{I}$) the velocity is given by

$$c^2 = (1+\beta(1+M'))(\gamma P^e/\beta . \rho_f^e) = (1+\beta(1+M'))(1-\beta)c_0^2.$$

$M'$ depends on the frequency, so the propagation is dispersive. Moreover, each of the diagonal terms $M'_{ii}$ are bounded below by $\beta/(1-\beta)$ (see Appendix C), so the celerity $c$ is greater than $c_0$,

$$c = c_0 \sqrt{(1+\beta(1+M'))(1-\beta)} \geqq c_0. \tag{C.16}$$

(C.16) may be compared with Crespo's result [C, 1969]: $c = c_0 \sqrt{(1+3 . \beta)(1-\beta)}$, which was obtained for weak concentrations and does not include the dispersion due to the capillary effect. In this case, the inertial coupling corresponds to $M' = 2$ which is the virtual mass effect of a single bubble in a liquid. For large concentrations, an analytical value can not be proposed since the bubble interactions are not negligible. However the calculation of $M'$ can be carried out using the variationnal formulation given in Appendix C.

### 4.4. Summary

Three behaviours have been obtained for fixed values of the dimensionless numbers. One can prove, as in [B & A, 1990], that there is a continuous evolution between all behaviours. Consequently, the validity range of each description can be delimited by intermediate values of the dimensionless numbers such as $\sqrt{\varepsilon} . \varepsilon^n$ ($n$ integer).

In Figure 2, we have drawn in the frequency-bubble diameter plane the domain of validity of the three cases, for an air- water mixture at equal concentration.

– The domain corresponding to the *case A*, is characterized by

$$\sqrt{\varepsilon^{-1}} \geqq N_v \geqq \sqrt{\varepsilon}, \qquad \sqrt{\varepsilon^{-1}} \geqq N_t \geqq \sqrt{\varepsilon}, \qquad \sqrt{\varepsilon} \geqq K_c . \beta/P^e \geqq \varepsilon\sqrt{\varepsilon}.$$

– The range of *case B* is given by

$$N_v \geqq \sqrt{\varepsilon^{-1}}, \qquad N_t \geqq \sqrt{\varepsilon^{-1}}, \qquad K_c . \beta/P^e \geqq \sqrt{\varepsilon}.$$

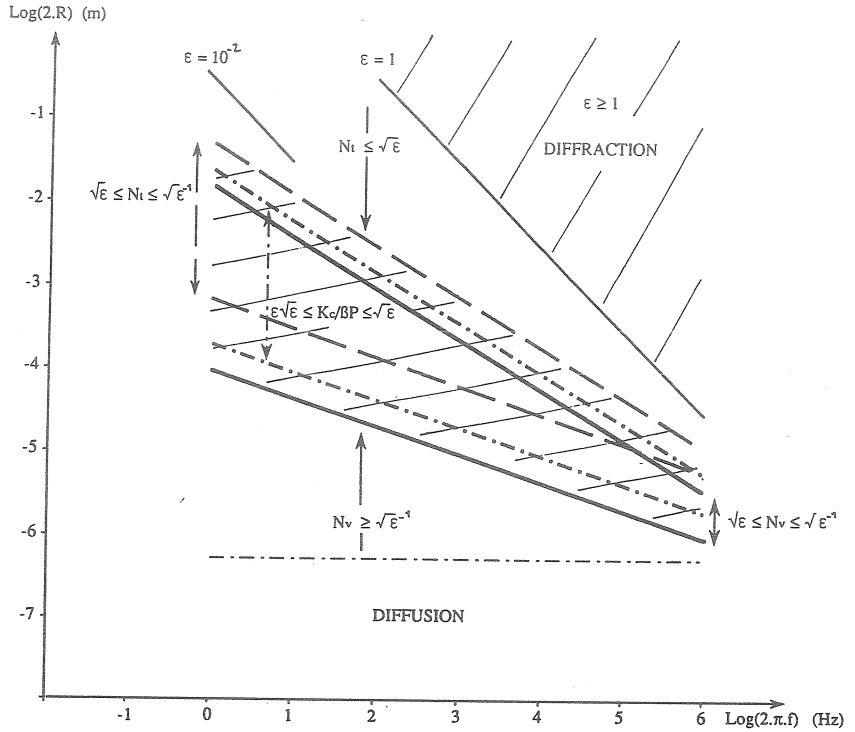


Fig. 2. – The validity ranges of the different behaviours of an air-water mixture at equal concentrations, according to the diameter of the bubbles and the wave frequency. $N_v \leqq \sqrt{\varepsilon^{-1}}$; $N_t \geqq \sqrt{\varepsilon}$: "Medium" bubble size and frequencies: Case A, Eqs. (A.15). $N_v \geqq \sqrt{\varepsilon^{-1}}$: "Small" bubble size: Case B, Eqs. (B.15). $N_t \leqq \sqrt{\varepsilon}$: "Large" bubble size, "high" frequencies: Case C, Eqs. (C.15).

– Finally *case C* is obtained when

$$N_v \leqq \sqrt{\varepsilon}, \qquad N_t \leqq \sqrt{\varepsilon}, \qquad K_c . \beta / P^e \leqq \varepsilon . \sqrt{\varepsilon}.$$

At a given frequency, the behaviours of cases A and C can be extended to mixtures with bubbles of different sizes, as long as we stay within the same region of validity. For the case B we have to define as many pressure fields as there are different diameters. Therefore, there are as many propagation modes as there are bubbles sizes (as for mixtures of two liquids [A & L, 1989]).

We recall that these descriptions cannot describe the diffraction effects which occur when $\varepsilon = 2\,R\,.\,\omega / c_0 \geqq 1$.

## 5. Conclusion

We have investigated the linear acoustics of bubbly fluids at large concentration, taking into account the bubble interactions, the viscosity, heat transfers and capillary effects. The results are valid for a large number of configurations, since they can be applied for mixtures constituted of liquid and gas with approximately the same mechanical and

thermal characteristics as water and air, with bubble sizes ranging from $10^{-8}$ to $10^{-2}$ m and for a wide frequency range (from 0.1 to $10^6$ Hz).

The small parameter $\varepsilon$ is used systematically in the homogenization process. It is used particulary to measure the ratios between various properties of the two phases and to evaluate the dimensionless numbers describing the physics on the local scale. In this manner, we are able to determine the validity conditions of the different models, and secondly to give their accuracy according to the bubbles' diameter and the frequency.

We show that three acoustic behaviours are possible.

– For small bubbles such that the rigidity of the capillary membrane is of the same order as the bulk incompressiblity, the capillary effects tend to decrease the sound celerity to such a point that the wave becomes diffusive. The viscous and thermal layers are large, therefore the bubbles do not move within in the fluid and the gas is under isothermal conditions.

– When the bubble size is of same order as the viscous and thermal layers, the acoustic waves are damped. The attenuation is due to the superposition of two effects which give rise to dampings of the same order of magnitude *i.e.* the displacements between the liquid and the gas, and the phase shift between pressure and gas density due to the thermal conduction.

– When the bubbles are large in comparison with the thermal and viscous layers, relative displacements occur leading to a virtual mass effect, and the gas is under adiabatic conditions. Finally, the capillary tension is so weak that bubbles oscillate asymmetrically.

## APPENDIX

### The homogenization process applied to bubbly fluids

Let us consider a medium made up of gas bubbles of identical diameter, periodically distribued in a fluid (*Fig.* 3). $\Omega$ is the volume of the periodic cell $\Gamma$ its boundary and $\Gamma_g$ the interface separating the two phases. $\Omega_f$ and $\Omega_g$ indicate the fluid and gas volume

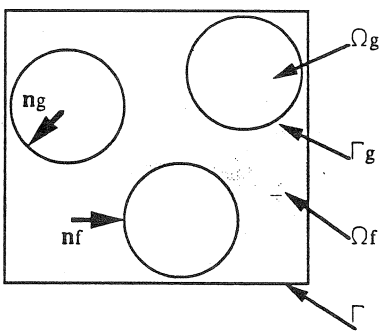


Fig. 3. – Geometric description of the gas-fluid mixture at finite concentration. The periodic cell of the mixture.

respectively $\mathbf{n}_f$, $\mathbf{n}_g$ their unit outward normal vector. The volumic concentration of the gas is defined by $\beta = |\Omega_g|/|\Omega|$, and that of the liquid by $1-\beta = |\Omega_f|/|\Omega|$.

In order to simplify the presentation, we study a cell containing one bubble only. We will see later that the results can easily be extended to cells containing several bubbles.

## APPENDIX A

### Case A : "Medium" bubble size and frequencies

### A1. The local description

The set of Eqs. (A.1-A.14) corresponding to the case A [$N_v = O(N_t) = O(1)$ and $K_c . \beta/P^e = O(\varepsilon)$] will be numbered (1A-14A). As described in Section 2, we introduce into the equations asymptotic expansions of each variable, and $\partial/\partial y$ is replaced by $\partial/\partial y + \varepsilon\,\partial/\partial x$. By collecting together terms with the same powers of $\varepsilon$ we obtain the first orders of Eqs. (1A-14A). (We denote them by using the same numbers with an additional index corresponding to the order.)

*In the liquid*

(2A-0) (2A-1) $\quad \sigma_f^0 = -P_f^0 . \mathbf{I}, \qquad \sigma_f^1 = -P_f^1 . \mathbf{I} + 2 . i\omega . \mu_f E_y(\mathbf{u}_f^0)$

(3A-0) $\quad \mathbf{grad}_y(P_f^0) = 0,$

(3A-1) $\quad \mathbf{grad}_x(P_f^0) + \mathbf{grad}_y(P_f^1) - i\omega . \mu_f (\Delta_y(\mathbf{u}_f^0) + \mathbf{grad}_y(\mathrm{div}_y(\mathbf{u}_f^0)) = \rho_f^e . \omega^2 . \mathbf{u}_f^0,$

(4A-0) (4A−1) $\quad \mathrm{div}_y(\mathbf{u}_f^0) = 0, \qquad \mathrm{div}_y(\mathbf{u}_f^1) + \mathrm{div}_x(\mathbf{u}_f^0) = 0,$

(5A-0) $\quad 0 = i\omega . (\rho_f^e . C_{pf} . T_f^1 - P_f^0).$

*In the gas*

(6A-0) $\quad P_g^0 = P^e . (\rho_g^0/\rho_g^e + T_g^0/T^e),$

(7A-0) (7A-1) $\quad \sigma_g^0 = -P_g^0 . \mathbf{I}, \qquad \sigma_g^1 = -P_g^1 . \mathbf{I},$

(8A-0) (8A-1) $\quad \mathbf{grad}_y(P_g^0) = 0, \qquad \mathbf{grad}_x(P_g^0) + \mathbf{grad}_y(P_g^1) = 0,$

(9A-0) (9A-1) $\quad \mathrm{div}_y(\mathbf{u}_g^0) = 0, \qquad \rho_g^0 + \rho_g^e . (\mathrm{div}_x(\mathbf{u}_g^0) + \mathrm{div}_y(\mathbf{u}_g^1)) = 0,$

(10A-0) $\quad \mathrm{div}_y(k_g . \mathbf{grad}_y(T_g^0)) = i\omega . (\rho_g^e . C_{pg} . T_g^0 - P_g^0).$

*At the interface*

(11A-0); (11A-1) $\quad \mathbf{u}_f^0 = \mathbf{u}_g^0, \qquad \mathbf{u}_f^1 = \mathbf{u}_g^1,$

(12A-0); (12A-1) $\quad (P_g^0 - P_f^0) . \mathbf{n} = 0, \qquad (\sigma_g^1 - \sigma_f^1) . \mathbf{n} = P_c^1 . \mathbf{n},$

(13A-0) $\quad T_g^0 = 0.$

### A2. The pressures

From the system (3A-0, 8A-0, 12A-0), we deduce that the pressure is constant over the period so that $P_f^0 = P_g^0 = P(x)$.

The pressure $P_g^1$ is determined by integrating equation (8A-1), which gives

(15A) $$P_g^1 = P_g'^1(x) - \mathbf{y} \cdot \mathbf{grad}_x(P).$$

### A3. The temperature and the gas density

Using expansions for the equilibrium pressure and temperature, Eq. (10A-0) becomes

$$\mathrm{div}_y(k_g \cdot \mathbf{grad}_y(T_g^0/T^e))/i\omega \cdot \rho_g^e \cdot C_{pg} - T_g^0/T^e = -P(1-1/\gamma)/P^e.$$

This, together with (13A-0), is a problem already solved in [A, 1983 (Sec. 4)] where it is shown that the temperature in the gas is given by

(16A) $$T_g^0 = P(x) \cdot g(\mathbf{y}) \cdot T^e(1-1/\gamma)/P^e,$$

where $g(\mathbf{y})$ is a complex function of the dimensionless frequency $\omega/\omega_t$ (with $\omega_t = d_g/R^2$). The density in the gas is related to the pressure by relation (6A-0) which gives

(17A) $$\rho_g^0/\rho_g^e = P/P^e - T_g^0/T^e, \quad \text{so that} \quad \rho_g^0 = P \cdot (1 - g(\mathbf{y})(1-1/\gamma))\,\rho_g^e/P^e.$$

### A4. The displacement fields

Since we assume that the variation of the curvature radius $r$ is $O(\varepsilon^2 \cdot R)$, while the displacement is $O(\varepsilon \cdot R)$, the modification of the radius of the bubble appears at the second order only. This requires that the interface does not change its form at the first order. Thus, on any point of the interface, the displacement $\mathbf{u}_f^0$ can be resolved into a constant translation over the period and a displacement tangential to the surface of the bubble:

(18A) $$\mathbf{u}_f^0 = \mathbf{u}_c(x) + \mathbf{u}' \qquad \text{with} \quad \mathbf{u}' \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_g.$$

Actually, $\mathbf{u}_c(x)$ is related to $\mathbf{u}_f^0$ by an integral condition since we have

$$\int_{\Gamma g} y_i(\mathbf{u}_f^0 \cdot \mathbf{n}_g)\,ds = \int_{\Gamma g} y_i(\mathbf{u}_c(x) + \mathbf{u}') \cdot \mathbf{n}_g\,ds = \mathbf{u}_{cj} \cdot \int_{\Gamma g} y_i \cdot n_{gj}\,ds = \mathbf{u}_{cj} \cdot \delta_{ij} |\Omega_g|,$$

which gives

(19A) $$\mathbf{u}_c(x) = |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{y}(\mathbf{u}_f^0 \cdot \mathbf{n}_g)\,ds.$$

## A4.1. A VARIATIONAL FORMULATION FOR THE LIQUID DISPLACEMENT FIELD

Eqs. (4A-0, 3A-1, 12A-1, 18A) govern the displacement field in $\Omega_f$. We study the structure of the solution by establishing a variational formulation. Let us consider the vector space W of complex test vectors $\mathbf{w}$ defined by

$$\{\mathbf{w} \text{ defined on } \Omega_f,\ \Omega\text{-periodic/div}(\mathbf{w})=0,\ \text{On } \Gamma_g : \mathbf{w}=\mathbf{w}_c(x)+\mathbf{w}';\ \mathbf{w}'.\mathbf{n}=0\},$$

associated with the hermitian product

$$(\mathbf{u},\ \mathbf{w})=\int_{\Omega f}(\mathbf{E}_y(\mathbf{u}):\mathbf{E}_y(\mathbf{w}^*)+\mathbf{u}.\mathbf{w}^*)\,dv,$$

(where $\mathbf{w}^*$ is the complex conjugate of $\mathbf{w}$).

When multiplying (3A-1) by any member $\mathbf{w}^*$ of W, and integrating over $\Omega_f$, we obtain

$$\int_{\Omega f}\mathbf{grad}_x(\mathrm{P}).\mathbf{w}^*\,dv-\int_{\Omega f}\mathbf{div}_y(\sigma_f^1).\mathbf{w}^*\,dv=\int_{\Omega f}\rho_f^e.\omega^2.\mathbf{u}_f^0.\mathbf{w}^*\,dv,$$

but

$$\int_{\Omega f}\mathbf{div}_y(\sigma_f^1).\mathbf{w}^*\,dv=\int_{\Omega f}(\sigma_{fij}^1\mathbf{w}_i^*)_{,j}\,dv-\int_{\Omega f}\sigma_{ij}^1.\mathbf{w}_{i,j}^*\,dv.$$

Due to the zero divergence of $\mathbf{w}^*$ and the symmetry of $\mathbf{E}_y$, the last term can be written in the form

$$-\int_{\Omega f}(-\mathrm{P}_f^1.\delta_{ij}+2.i\omega.\mu_f\mathbf{E}_y(\mathbf{u}_f^0)_{ij})\mathbf{w}_{i,j}^*\,dv=-\int_{\Omega f}i\omega.\mu_f\mathbf{E}_y(\mathbf{u}_f^0)_{ij}\mathbf{E}_y(\mathbf{w}^*)_{ij}\,dv.$$

Using the divergence theorem, we obtain

$$\int_{\Omega f}(\sigma_{fij}^1\mathbf{w}^*{}_i)_{,j}\,dv=\int_{\Gamma\cup\Gamma g}(\sigma_{fij}^1\mathbf{w}_i^*).\mathbf{n}_j\,ds,$$

which, because of (12A-1) and (15A) and the periodicity, implies

$$-\int_{\Gamma g}(\mathrm{P}_g^1+\mathrm{P}_c^1)\mathbf{w}_j^*.\mathbf{n}_j\,ds$$
$$=-\int_{\Gamma g}\mathrm{P}_g'^1(x)\mathbf{w}_j^*.\mathbf{n}_j\,ds+\int_{\Gamma g}(\mathbf{y}.\mathbf{grad}_x(\mathrm{P}))\mathbf{w}_j^*.\mathbf{n}_j\,ds-\mathbf{w}_c(x).\int_{\Gamma g}\mathrm{P}_c^1.\mathbf{n}_j\,ds.$$

The kinematic condition (18A) implies that the first integral is zero. By using the momentum balance applied to the zero mass interface, the last integral also disappears. Finally, putting

$$\langle\mathbf{u}_f^0,\ \mathbf{w}\rangle=-(i\mu_f/\rho_f^e.\omega)\int_{\Omega f}\mathbf{E}_y(\mathbf{u}_f^0)_{ij}\mathbf{E}_y(\mathbf{w}^*)_{ij}\,dv+\int_{\Omega f}\mathbf{u}_f^0.\mathbf{w}^*\,dv,$$

the variational formulation becomes

$$(20A) \qquad \forall \mathbf{w} \in W, \qquad \langle \mathbf{u}_f^0, \mathbf{w} \rangle = (\mathbf{grad}_x(P)/\rho_f^e . \omega^2)\left( \int_{\Omega f} \mathbf{w}^* \, dv - \int_{\Gamma g} \mathbf{y}(\mathbf{w}^* \mathbf{n}) \, ds \right).$$

### A4.2. The fluid displacement field

The variational formulation (20A) displays the linearity of the problem, and the Lax-Milgram lemma ensures the existence and the uniqueness of $\mathbf{u}_f^0$ which depends linearly on the pressure gradient. It is convenient to resolve the solution into two terms.

– The first terms is due to the forcing term in the fluid volume, *i.e.* the first term in the right hand member of (20A). The corresponding solution is obviously the constant vector

$$\mathbf{grad}_x(P)/\rho_f^e . \omega^2.$$

– The second term is due to the stresses applied on the surface of the bubble *i.e.* the last term of (20A). It leads to solutions of the form:

$$\mathbf{m} . \mathbf{grad}_x(P)/\rho_f^e . \omega^2.$$

The tensor $\mathbf{m}$ is made up of the complex fields $\mathbf{m}^p$ which depend on the local variable and the dimensionless frequency $\omega/\omega_v$ (with $\omega_v = \nu_f/R^2$) since they are solutions of

$$(21A) \qquad \forall \mathbf{w} \in W, \qquad \langle \mathbf{u}, \mathbf{w} \rangle = -(\mathbf{grad}_x(P)/\rho_f^e . \omega^2)\left( \int_{\Gamma g} \mathbf{y}(\mathbf{w}^* . n) \, ds \right),$$

when

$$(\mathbf{grad}_x(P)/\rho_f^e . \omega^2)_i = \delta_{ip}.$$

In conclusion we have

$$(22A) \qquad \mathbf{u}_f^0 = (\mathbf{grad}_x(P)/\rho_f^e . \omega^2)(\mathbf{I} + \mathbf{m}).$$

### A4.3. The average displacement field in the gas

As the bubbles are not connected, the determination of $\mathbf{u}_f^0$ allows us to find directly the average displacement in the gas. Taking into account equations (9A-0, 11A-0), we integrate over $\Omega_g$ the identity $(\mathbf{u}_{gk}^0 y_i)_{,k} = \mathrm{div}_y(\mathbf{u}_g^0) . y_i + \delta_{ik} . \mathbf{u}_{gk}^0$, to give

$$\int_{\Omega g} (\mathbf{u}_{gk}^0 y_i)_{,k} \, dv = \int_{\Omega g} \mathbf{u}_{gi}^0 \, dv = \int_{\Gamma g} (\mathbf{u}_{gk}^0 . y_i) . \mathbf{n}_{gk} \, ds = \int_{\Omega g} y_i (\mathbf{u}_g^0 . \mathbf{n}_g) \, ds.$$

Using expression (19A) the average displacement in the gas is therefore given by

$$(23A) \qquad \mathbf{U}_g = |\Omega_g|^{-1} \int_{\Omega g} \mathbf{u}_g^0 . \, dv = \mathbf{u}_c(x).$$

## A5. The balance equations

### A5.1. The momentum balance

On integrating the momentum balances of the two phases (3A-1) and (8A-1) over their respective domain of definition and adding term by term, we find

$$\int_{\Omega} \mathbf{grad}_x(\mathrm{P})\, dv - \int_{\Gamma \cup \Gamma g} \sigma_f^1 . \mathbf{n}_f\, ds + \int_{\Gamma g} \mathrm{P}_g^1 . \mathbf{n}_g\, ds = \int_{\Omega f} \rho_f^e . \omega^2 . \mathbf{u}_f^0\, dv.$$

The two surface integrals cancel out because of the periodicity, stress continuity (12A-1) and the equilibrium of the interface $\left(\text{since they have a zero mass } i.e. \int_{\Gamma g} \mathrm{P}_c^1 . \mathbf{n}_g . ds = 0\right)$. Therefore we obtain the macroscopic momentum balance

$$\mathbf{U}_f = |\Omega_f|^{-1} \int_{\Omega f} \mathbf{u}_f^0\, dv = (1/\rho_f^e . \omega^2)\,(\mathbf{grad}_x \mathrm{P} + (\beta/(1-\beta)\,\mathbf{grad}_x \mathrm{P}).$$

By comparing with expression (22A) of $\mathbf{u}_f^0$, we obtain

$$|\Omega_f|^{-1} \int_{\Omega f} \mathbf{m}\, dv = \beta/(1-\beta)\,\mathbf{I}. \tag{24A}$$

The macroscopic momentum balance of the gas is given by (23A, 19A, 22A):

$$\mathbf{U}_g = |\Omega_g|^{-1} \int_{\Omega g} \mathbf{u}_g^0\, dv = (\mathbf{grad}_x(\mathrm{P})/\rho_f^e . \omega^2)\, |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{y} . (\mathbf{I} + \mathbf{m})\, \mathbf{n}_g\, ds$$

*i.e.*

$$\mathbf{U}_g = (1/\rho_f^e . \omega^2)\,(\mathbf{I} + \mathbf{M}) . \mathbf{grad}_x(\mathrm{P}), \qquad \text{with} \quad \mathbf{M} = |\Omega_g|^{-1} \int_{\Gamma g} (\mathbf{y} . \mathbf{m})\, \mathbf{n}_g\, ds.$$

### A5.2. The volume balance

Using the expression (17A) for the gas density fluctuation, and integrating (4A-1) and (9A-1) over $\Omega_f$ and $\Omega_g$ respectively, we get after addition,

$$\int_{\Omega f} \mathrm{div}_x(\mathbf{u}_f^0)\, dv + \int_{\Omega g} \mathrm{div}_x(\mathbf{u}_g^0)\, dv + \int_{\Gamma g} \mathbf{u}_f^1 . \mathbf{n}_f\, ds + \int_{\Gamma g} \mathbf{u}_g^1 . \mathbf{n}_g\, ds$$

$$= -\int_{\Omega g} \mathrm{P} . (1 - g(\mathbf{y})\,(1 - 1/\gamma))/\mathrm{P}^e\, dv.$$

As a result of (11A-1) and the periodicity, the surface integrals cancel out. After reversing the order of the $y$-integration and the $x$-differentiation, there exists only one macroscopic

volume balance:

$$\mathrm{div}_x\left(\int_{\Omega f}\mathbf{u}_f^0\,.\,dv\right)+\mathrm{div}_x\left(\int_{\Omega g}\mathbf{u}_g^0\,.\,dv\right)=-|\Omega_g|\,.\,\mathrm{P}\,(1-\mathrm{G}\,(1-1/\gamma))/\mathrm{P}^e,$$

*i.e.*

$$(1-\beta)\,.\,\mathrm{div}_x\,(\mathbf{U}_f)+\beta\,.\,\mathrm{div}_x\,(\mathbf{U}_g)=\beta\,.\,\mathrm{P}\,(-1+\mathrm{G}\,(1-1/\gamma))/\mathrm{P}^e,$$

with

$$\mathrm{G}=|\Omega_g|^{-1}\int_{\Omega g}g\,(\mathbf{y})\,dv.$$

## A6. Macroscopic description

Finally, we obtain at the macroscopic level a medium governed by the constitutive equations below, where the index $_x$ has been omitted. G and M are, respectively, a complex scalar function of the dimensionless frequency $\omega/\omega_t$ and a complex symmetric tensor depending on $\omega/\omega_v$.

$$\text{(25A)}\qquad \begin{cases} (1-\beta)\,.\,\mathbf{U}_f=(1/\rho_f\,.\,\omega^2)\,\mathbf{grad}\,(\mathrm{P}), \\ \beta\,.\,\mathbf{U}_g=(\beta/\rho^e\,.\,\omega^2)\,(\mathbf{I}+\mathbf{M})\,\mathbf{grad}\,(\mathrm{P}), \\ \mathrm{div}\,((1-\beta)\,.\,\mathbf{U}_f)+\mathrm{div}\,(\beta\,.\,\mathbf{U}_g)=\beta\,(-1+\mathrm{G}\,(1-1/\gamma))\,\mathrm{P}/\mathrm{P}^e. \end{cases}$$

### A6.1. Some properties of the function

In [A, 1983] it is show that $0\leqq\mathrm{Re}\,(\mathrm{G})\leqq 1$; $0\leqq\mathrm{Im}\,(\mathrm{G})\leqq 1$. Moreover it is shown that when $\omega/\omega_t\to 0$, $\mathrm{G}\to 0$ with $\mathrm{Re}\,(\mathrm{G})=\mathrm{O}\,(\omega^2)$ and $\mathrm{Im}\,(\mathrm{G})=\mathrm{O}\,(\omega)$. Also when $\omega/\omega_t\to\infty$, $\mathrm{G}\to 1$.

For spherical bubbles G can be expressed analytical in the following form.

$$\mathrm{G}=1+(3/i\,\omega_t^*)\,(1-\sqrt{i\,\omega_t^*}\,.\coth(\sqrt{i\,\omega_t^*})),\qquad \omega_t^*=\omega/\omega_t.$$

### A6.2. Properties of the tensor M

The components of tensor **M** are given by $\mathrm{M}_{ij}=|\Omega_g|^{-1}\int_{\Gamma g}y_j\,.\,m_k^i\,.\,n_{gk}\,ds$. Let us consider the particular solution $\mathbf{m}^p$ of (21A). Putting $\mathbf{w}^*=\mathbf{m}^r$ we have

$$\mathrm{M}_{rp}=|\Omega_g|^{-1}\,\langle\,\mathbf{m}^p,\,\mathbf{m}^{r*}\,\rangle.$$

From the symmetry of the right hand side, we deduce the symmetry of $\mathbf{M}$. The real and imaginary parts of $M_{ii}$ (without summation on $i$) are given by

$$\mathrm{Re}\,(M_{ii}^{*}) = (M_{ii} + M_{ii}^{*})/2 = |\Omega_g|^{-1}\left(\int_{\Omega f} \mathbf{m}^i . \mathbf{m}^{i*}\, dv\right) = |\Omega_g|^{-1}\left(\int_{\Omega f} |\mathbf{m}^{i\,|\,2}\, dv\right) \geqq 0,$$

$$\mathrm{Im}\,(M_{ii}^{*}) = (M_{ii} - M_{ii}^{*})/2\, i = |\Omega_g|^{-1}\,(i\,\mu_f/\rho_f^e . \omega) \int_{\Omega f} \mathbf{E}_y(\mathbf{m}^i)_{jk}\, \mathbf{E}_y(\mathbf{m}^{i*})_{jk}\, dv) \geqq 0.$$

Using the Schwartz inequality and (24A) we get the lower bound

(26A) $$\mathrm{Re}\,(M_{ii}^{*}) = |\Omega_g|^{-1}\left(\int_{\Omega f} |\mathbf{m}^i|^2\, dv\right) \geqq \beta/(1-\beta) \quad \text{(no summation on } i\text{)}.$$

Lastly, from Eq. (21A) it is clear that $\mathbf{m}$ tends to a purely imaginary value when $\omega/\omega_v$ tends to zero, and $\mathbf{m}$ tends to a real value when $\omega/\omega_v$ tends to infinity.

### A6.3. Comments

The hypothesis of the presence of only one bubble in the period can be dispensed with, by introducing a kinematic condition for each bubble. In this way, the variational formulation remains similar and there is no modification of the macroscopic description.

## APPENDIX B

### Case B: "small" bubble size

### B1. The local description

The equations to be treated in the case B [$N_v = O(N_t) = O(\varepsilon^{-1})$, $K_c . \beta/P^e = O(1)$] are the same as in Appendix A except for those below which are now indexed with B.

*In the liquid*

(2B-0) $$\sigma_f^0 = -P_f^0 . \mathbf{I} + 2 . i\omega . \mu_f\, \mathbf{E}_y(\mathbf{u}_f^0)$$

(2B-1) $$\sigma_f^1 = -P_f^1 . \mathbf{I} + 2 . i\omega . \mu_f\, (\mathbf{E}_x(\mathbf{u}_f^0) + \mathbf{E}_y(\mathbf{u}_f^1)),$$

(3B-0) $$\mathbf{grad}_y(P_f^0) - i\omega . \mu_f\, (\Delta_y(\mathbf{u}_f^0) + \mathbf{grad}_y(\mathrm{div}_y(\mathbf{u}_f^0))) = 0,$$

(3B-1) $$-i\omega . \mu_f\, ((\Delta_{yx}(\mathbf{u}_f^0) + \Delta_y(\mathbf{u}_f^1 + \mathbf{grad}_y(\mathrm{div}_y(\mathbf{u}_f^1)) + \mathbf{grad}_y(\mathrm{div}_x(\mathbf{u}_f^0)) + \mathbf{grad}_x(P_f^0) + \mathbf{grad}_y(P_f^1) = \rho_f^e . \omega^2 . \mathbf{u}_f^0.$$

*In the gas*

(6B-0) $$P_g^0 = P^e . \rho_g^0/\rho_g^e,$$

(7B-0) (7B-1) $$\sigma_g^0 = -P_g^0 . \mathbf{I}, \qquad \sigma_g^1 = -P_g^1 . \mathbf{I} + 2 . i\omega . \mu_g\, \mathbf{E}_y(\mathbf{u}_g^0),$$

(8B-0) $$\mathbf{grad}_y(P_g^0)=0,$$

(8B-1) $$\mathbf{grad}_x(P_g^0)+\mathbf{grad}_y(P_g^1)-i\omega.\mu_g(\Delta_y(\mathbf{u}_g^0)+\mathbf{grad}_y(\mathrm{div}_y(\mathbf{u}_g^0)))=0.$$

*At the interfaces*

(12B-0) (12B-1) $$(\sigma_g^0-\sigma_f^0).\mathbf{n}=P_c^0.\mathbf{n}, \qquad (\sigma_g^1-\sigma_f^1).\mathbf{n}=P_c^1.\mathbf{n}.$$

## B2. The pressure and the density of the gas

Equation (8B-0) gives a constant gas pressure at the first order: $P_g^0=P_g(x)$.

Equation (6B-0) leads to a constant perturbation of density on the bubble:

(27B) $$\rho_g^0=\rho_g^e P_g/P^e$$

Notice that there is no thermal effect at the first order since the gas is under isothermal conditions. Therefore the investigation of the acoustics of the mixture does not need to study the thermal conduction any further.

## B3. Displacement and pressure in the liquid

Using the same idea as in Appendix A, we must add to Eqs. (4A-0, 3B-0, 12B-0), the kinematic condition (18A) expressing the invariance of the form of the bubble at the first order. It is easy to show that the variational formulation of this problem is

$$\forall \mathbf{w}\in W, \qquad -(i\mu_f.\omega)\int_{\Omega f}\mathbf{E}_y(\mathbf{u}_f^0)_{ij}\,\mathbf{E}_y(\mathbf{w}^*)_{ij}\,dv=0,$$

from which we get $\mathbf{u}_f^0=\mathbf{U}(x)$. Then $P_f^0=P_f(x)$ and with (12B-0) $P_c^0=P_c(x)$.

As in the previous case, we can express the fluid displacement in the gas volume by

$$\mathbf{U}_g=|\Omega_g|^{-1}\int_{\Omega g}\mathbf{u}_g^0.dv=\mathbf{u}_c(x)=\mathbf{U}(x).$$

## B4. Balance equations

### B4.1. Momentum balance

Integrating the momentum balances (3B-1) and (8B-1) over their respective domains leads to

$$\int_{\Omega f}\mathbf{grad}_x(P_f)\,dv+\int_{\Omega_g}\mathbf{grad}_x(P_g)\,dv-\int_{\Gamma\cup\Gamma g}\sigma_f^1.\mathbf{n}_f\,ds-\int_{\Gamma g}\sigma_g^1.\mathbf{n}_g\,ds=\int_{\Omega f}\rho_f^e.\omega^2.\mathbf{U}\,dv.$$

The two surface integrals cancel out as before (*see* Appendix A). We are left with

$$\mathbf{grad}_x(P_f) + \beta/(1-\beta)\,\mathbf{grad}_x(P_g) = (\rho_f^e \,.\, \omega^2)\,\mathbf{U}.$$

B4.2. MASS BALANCE

With (6B-0), Eq. (9A-1) expressing the local mass balances in the gas is as follows

$$P_g/P^e + (\mathrm{div}_y(\mathbf{u}_g^1) + \mathrm{div}_x(\mathbf{u}_g^0)) = 0 \quad \text{in } \Omega_g.$$

Separately integrating (4A-1) and (9A-1) over their own domains we obtain

$$\int_{\Gamma \cup \Gamma g} \mathbf{u}_f^1 \,.\, n_f \, ds + |\Omega_f| \,.\, \mathrm{div}_x(\mathbf{U}) = 0.$$

So

$$\int_{\Gamma g} \mathbf{u}_g^1 \,.\, n_g \, ds + \mathrm{div}_x\left[\int_{\Omega g} \mathbf{u}_g^0 \, dv\right] = -|\Omega_g| \,.\, P_g/P^e.$$

But the capillary pressure is constant over all the interface and consequently the curvature is a constant at any point of the capillary membrane. That means that the first significant deformation occurs in a spherical mode. Thus, we have

$$-\mathbf{u}_f^1 \,.\, \mathbf{n}_f = \mathbf{u}_g^1 \,.\, \mathbf{n}_g = r^1 = P_c(x)\,(R^2/2\,\sigma),$$

so that $|\Omega_g|^{-1} \int_\Gamma \mathbf{u}_g^1 \,.\, \mathbf{n}_g \, ds = P_c(x)/K_c$ with $K_c = 2\,\sigma/3\,R$.

And as $|\Omega_g|^{-1} \int_{\Omega g} \mathbf{u}_g^0 \,.\, dv = \mathbf{U}$,

we successively obtain

$$\mathrm{div}_x(\mathbf{U}) = (\beta/1-\beta)\,P_c/K_c = (\beta/1-\beta)\,.\,(P_f - P_g)/K_c$$

and

$$\mathrm{div}_x(\mathbf{U}) = -P_c/K_c - P_g/P^e = -P_f/K_c - P_g\,(1/P^e - 1/K_c)$$

*i.e.*

$$P_g = -(P^e/\beta)\,\mathrm{div}_x(\mathbf{U}), \qquad P_f = -(1-(1-\beta)\,K_c/P^e)\,(P^e/\beta)\,\mathrm{div}_x(\mathbf{U}).$$

**B5. Macroscopic description**

Macroscopically the mixture behaves like a medium which satisfies the equations:

(28B)
$$P_g = -(P^e/\beta)\,\mathrm{div}_x(\mathbf{U})$$
$$P_f = -(1-(1-\beta)\,K_c/P^e)\,(P^e/\beta)\,\mathrm{div}_x(\mathbf{U}), \qquad \text{with} \quad K_c = 2\,\sigma/3\,R,$$

$$\mathbf{grad}_x(P_f)+(\beta/1-\beta)\,\mathbf{grad}_x(P_g)=(\rho_f^e.\omega^2)\,\mathbf{U}.$$

We can verify that the presence of N bubbles $\Omega_{gi}$ $(i=1, \ldots, N)$ of the same diameter does not change the results, if we put $P_g=|\Omega_g|^{-1}.\sum_{i=1}^{N}\int_{\Omega gi} P_{gi}(x)\,dv$.

## APPENDIX C

### Case C : "Large" bubble size and frequencies

### C1. Local description

For the case C ($N_v=O(N_t)=O(\varepsilon)$ and $K_c.\beta/P^e=O(\varepsilon^2)$) the set of equations at different orders used in the homogenization process are those of Appendix A, except for the following which are now indexed with C.

*For the liquid*

(2C-0) (2C-1) $\qquad \sigma_f^0=-P_f^0.\mathbf{I}, \qquad \sigma_f^1=-P_f^1.\mathbf{I},$

(3C-0) (3C-1) $\qquad \mathbf{grad}_y(P_f^0)=0, \qquad \mathbf{grad}_x(P_f^0)+\mathbf{grad}_y(P_f^1)=\rho_f^e.\omega^2.\mathbf{u}_f^0.$

*For the gas*

(10C-0) $\qquad 0=i\omega.(\rho_g^e.C_{pg}.T_g^0-P_g^0).$

### C2. The pressure, temperature and density in the gas

Using (3C-0), (8C-0) and (12A-0), we get $P_f^0=P_g^0=P(x)$.

The gas pressure at the next order is given by (8C-1) $P_g^1=-y.\mathbf{grad}_x(P)+P_g'^1(x)$.

The temperature is given by (10C-0) which leads to $T_g=P/\rho_g^e.C_{pg}$.

Therefore the oscillation of the gas density, which is given by Eq. (6A-0),

$$\rho_g/\rho_g^e=P/P^e-T_g/T^e=(1-P^e/T^e.\rho_g^e.C_{pg})\,P/P^e$$

is connected to the pressure by the adiabatic law $\rho_g/\rho_g^e=P/\gamma P^e$.

### C3. The displacement field

The displacements are governed by Eqs. (4A-0, 3C-1, 11A-0, 12A-1, 9A-0). We study this differential system using its variational formulation. With this aim, let us take the

scalar product of (3C-1) with any test field of the vectorial space $W_1$ defined by

$$W_1 = \{ \mathbf{w} \text{ defined over } \Omega_f / \mathbf{w} \text{ real, } \Omega\text{-periodic, } \mathrm{div}_y(\mathbf{w}) = 0 \}$$

and then integrate over $\Omega_f$,

$$\int_{\Omega f} \mathbf{grad}_x(\mathrm{P}) \,.\, \mathbf{w}\, dv + \int_{\Omega f} \mathbf{grad}_y(\mathrm{P}_f^1) \,.\, \mathbf{w}\, dv = \int_{\Omega f} \rho_f^e \,.\, \omega^2 \,.\, \mathbf{u}_f^0 \,.\, \mathbf{w}\, dv.$$

When making use of (12A-1) and the periodicity of $\mathrm{P}_f^1$, we have

$$\int_{\Omega f} \mathbf{grad}_y(\mathrm{P}_f^1) \,.\, \mathbf{w}\, dv = \int_{\Omega f} \mathrm{div}_y(\mathrm{P}_y^1 \,.\, \mathbf{w})\, dv - \int_{\Omega f} \mathrm{P}^1 \,.\, \mathrm{div}_y(\mathbf{w})\, dv$$

$$= \int_{\Gamma \cup \Gamma g} \mathrm{P}_f^1 \,.\, \mathbf{w} \,.\, \mathbf{n}_f\, ds = \int_{\Gamma g} (\mathrm{P}_g^1 + \mathrm{P}_c^1) \,.\, \mathbf{w} \,.\, \mathbf{n}_f\, ds.$$

With the expression for $\mathrm{P}_g^1$, it becomes

$$\int_{\Gamma g} (\mathrm{P}_g^1 + \mathrm{P}_c^1) \,.\, \mathbf{w} \,.\, \mathbf{n}_f\, ds = \int_{\Gamma g} \mathrm{P}_g'^1(x)\, \mathbf{w} \,.\, \mathbf{n}_f\, ds - \int_{\Gamma g} (\mathbf{y} \,.\, \mathbf{grad}_x(\mathrm{P}))\, \mathbf{w} \,.\, \mathbf{n}_f\, ds + \int_{\Gamma g} \mathrm{P}_c^1 \,.\, \mathbf{w} \,.\, \mathbf{n}_f\, ds.$$

Finally, by using the zero divergence of $\mathbf{w}$, the variational form becomes: $\forall \mathbf{w} \in W_1$,

$$\mathbf{grad}_x(\mathrm{P}) \left( \int_{\Omega f} \mathbf{w}\, dv - \int_{\Gamma g} \mathbf{y}\, (\mathbf{w} \,.\, \mathbf{n}_f)\, ds \right) = \rho_f^e \,.\, \omega^2 \int_{\Omega f} \mathbf{u}_f^0 \,.\, \mathbf{w}\, dv - \int_{\Gamma g} \mathrm{P}_c^1 \,.\, \mathbf{w} \,.\, \mathbf{n}_f\, ds.$$

Since here an asymmetrical deformation may appear, the kinematic condition (18A) used up to now is no longer satisfied, and we have to express $\mathrm{P}_c^1$ as a function of $\mathbf{u}_f^0$.

### C3.1. The capillary tension

The motion at any point on the interface can be resolved into a bulk displacement $\mathbf{u}_c$, corresponding to the displacement of the centre of gravity of the bubble, plus the normal and tangential displacements $\mathbf{u}_n$ and $\mathbf{u}_t$: $\mathbf{u}_f^0 = \mathbf{u}_c(x) + \mathbf{u}_n + \mathbf{u}_t$.

As in Appendix A the displacement $\mathbf{u}_c$ is equal to $\mathbf{u}_c(x) = |\Omega_g|^{-1} \int_{\Omega g} \mathbf{u}_g^0\, dv$ and because of the initial sphericity of the bubble $\mathbf{y} = -\mathrm{R} \,.\, \mathbf{n}_f$, so that

$$\mathbf{u}_c(x) = |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{y}\, (\mathbf{u}_f^0 \,.\, \mathbf{n}_g)\, ds = \mathrm{R} \,.\, |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{n}_f\, (\mathbf{u}_f^0 \,.\, \mathbf{n}_f)\, ds. \tag{29C}$$

At first order, the capillary pressure is due only to the normal component $\mathbf{u}_n$,

$$\mathrm{P}_c^1 = -(2\sigma/\mathrm{R}^2)\, \mathbf{u}_n \,.\, \mathbf{n}_f = -(2\sigma/\mathrm{R}^2)\, (\mathbf{u}_f^0 - \mathbf{u}_c)\, \mathbf{n}_f. \tag{30C}$$

Thus

$$\int_{\Gamma g} \mathrm{P}_c^1 . \mathbf{w} . \mathbf{n}_f \, ds = \int_{\Gamma g} -(2\sigma/\mathrm{R}^2)((\mathbf{u}_f^0 - \mathbf{u}_c) . \mathbf{n}_f) \mathbf{w} . \mathbf{n}_f \, ds$$

$$= -(2\sigma/\mathrm{R}^2)\left( \int_{\Gamma g} (\mathbf{u}_f^0 . \mathbf{n}_f)(\mathbf{w} . \mathbf{n}_f) \, ds - \int_{\Gamma g} (\mathbf{u}_c . \mathbf{n}_f)(\mathbf{w} . \mathbf{n}_f) \, ds \right)$$

and by using expression (29C) for $\mathbf{u}_c(x)$, we obtain

$$\int_{\Gamma g} \mathrm{P}_c^1 . \mathbf{w} . \mathbf{n}_f \, ds$$

$$= -(2\sigma/\mathrm{R}^2)\left( \int_{\Gamma g} (\mathbf{u}_f^0 . \mathbf{n}_f)(\mathbf{w} . \mathbf{n}_f) \, ds - \mathrm{R} . |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{n}_f (\mathbf{u}_f^0 . \mathbf{n}_f) \, ds \int_{\Gamma g} \mathbf{n}_f (\mathbf{w} . \mathbf{n}_f) \, ds \right)$$

Then, putting

$$\langle \mathbf{u}, \mathbf{w} \rangle_1 = \int_{\Omega f} \mathbf{u}_f^0 . \mathbf{w} \, dv + (2\sigma/\rho_f^e \omega^2 \mathrm{R}^2)$$

$$\times \left( \int_{\Gamma g} (\mathbf{u}_f^0 . \mathbf{n}_f)(\mathbf{w} . \mathbf{n}_f) \, ds - \mathrm{R} |\Omega_g|^{-1} \int_{\Gamma g} \mathbf{n}_f (\mathbf{u}_g^0 . \mathbf{n}_f) \, ds . \int_{\Gamma g} \mathbf{n}_f (\mathbf{w} . \mathbf{n}_f) \, ds \right),$$

the variational formulation becomes

$$\forall \mathbf{w} \in \mathrm{W}_1, \quad \langle \mathbf{u}_f^0, \mathbf{w} \rangle_1 = (1/\rho_f^e \omega^2) \mathbf{grad}_x(\mathrm{P}) \left( \int_{\Omega f} \mathbf{w} \, dv - \int_{\Gamma g} \mathbf{y} . \mathbf{w} . \mathbf{n}_f \, ds \right).$$

In this form the linearity of the problem clearly appears. However it remains necessary for us to prove that $\langle \mathbf{u}_f^0, \mathbf{u}_f^0 \rangle_1$ is strictly positive, to be in a position to apply the Lax-Milgram lemma.

C3.2. $\langle \ , \ \rangle_1$ IS A SCALAR PRODUCT

Obviously, the term $\int_{\Omega f} \mathbf{u}_f^0 . \mathbf{u}_f^0 \, dv$ meets this condition. For the other terms of $\langle \mathbf{u}_f^0, \mathbf{u}_f^0 \rangle_1$ we use expression (29C) of $\mathrm{P}_c^1$ to obtain

$$-\int_{\Gamma g} \mathrm{P}_c^1 . \mathbf{u}_f^0 . \mathbf{n}_f \, ds = (2\sigma/\mathrm{R}^2) \int_{\Gamma g} (\mathbf{u}_n . \mathbf{n}_f)(\mathbf{u}_n + \mathbf{u}_c) . \mathbf{n}_f \, ds$$

$$= (2\sigma/\mathrm{R}^2)\left( \int_{\Gamma g} (\mathbf{u}_n . \mathbf{n}_f)^2 \, ds + \mathbf{u}_c . \int_{\Gamma g} (\mathbf{u}_n . \mathbf{n}_f) \mathbf{n}_f \, ds \right).$$

But, as above,

$$0 = \int_{\Gamma g} \mathrm{P}_c^1 \mathbf{n}_f \, ds = -(2\sigma/\mathrm{R}^2) \int_{\Gamma g} (\mathbf{u}_n . \mathbf{n}_f) \mathbf{n}_f \, ds,$$

so

$$-\int_{\Gamma g} P_c^1 . \mathbf{u}_f^0 . \mathbf{n}_f \, ds = \int_{\Gamma g} (\mathbf{u}_n . \mathbf{n}_f)^2 \, ds \geqq 0.$$

Since $\langle \ , \ \rangle_1$ is a symmetric definite positive bilinear form, use now the Lax-Milgramm lemma to deduce the existence and the uniqueness of the solution. Consequently

$$\mathbf{u}_f^0 = (\mathbf{grad}_x (P)/\rho_f^e . \omega^2)(\mathbf{I} + \mathbf{m}'),$$

where $\mathbf{m}'$ is a second order tensor constructed from the solutions $\mathbf{m}'^p$ of the problems

$$\forall \mathbf{m} \in W_1, \quad \langle \mathbf{u}, \mathbf{w} \rangle_1 = -(\mathbf{grad}_x (P)/\rho_f^e . \omega^2) \left( \int_{\Gamma g} \mathbf{y} (\mathbf{w} . \mathbf{n}_f) \, ds \right)$$

when

$$(\mathbf{grad}_x (P)/\rho_f^e . \omega^2)_i = \delta_{ip}.$$

These particular solutions $\mathbf{m}'^p$ depend only on the dimensionless frequency

$$(2\sigma/\rho_f^e \omega^2 . R^2) |\Gamma_g| . |\Omega_g|^{-1} = (1/2)(\omega_r/\omega)^2 \qquad \text{where} \quad (\omega_r)^2 = 12 . \sigma/\rho^e{}_f . R^3.$$

## C4. The balance equations

They are obtained in the same mammer as in Appendix A:

*Momentum balance:*

$$\mathbf{grad}_x P = (\rho_f^e . \omega^2) \mathbf{U}_f \qquad \text{where} \quad \mathbf{U}_f = |\Omega_f|^{-1} \int_{\Omega f} \mathbf{u}_f^0 \, dv,$$

which again proves that

$$(1-\beta) . |\Omega_f|^{-1} \int_{\Omega f} (\mathbf{I} + \mathbf{m}') . dv = \mathbf{I} \quad \text{so} \quad |\Omega_f|^{-1} \int_{\Omega f} \mathbf{m}' \, dv = (\beta/1-\beta) \mathbf{I}.$$

*Volume balance:*

$$\operatorname{div}((1-\beta) . \mathbf{U}_f) + \operatorname{div}(\beta . \mathbf{U}_g) = -(\beta/\gamma P^e) P$$

with $\mathbf{U}_g = |\Omega_g|^{-1} \int_{\Omega g} \mathbf{u}_g^0 \, dv.$

## C5. Macroscopic description

Finally we get the constitutive equations of the homogenized medium:

$$\text{(31C)} \qquad \begin{cases} (1-\beta).\mathbf{U}_f = -(1/\rho_f^e.\omega^2)\,\mathbf{grad}\,(\mathrm{P}) \\ \beta.\mathbf{U}_g = -(\beta/\rho_f^e.\omega^2)(\mathbf{I}+\mathbf{M}')\,\mathbf{grad}\,(\mathrm{P}), \\ \mathrm{div}\,((1-\beta).\mathbf{U}_f)+\mathrm{div}\,(\beta.\mathbf{U}_g)=(\beta/\gamma\,\mathrm{P}^e)\,\mathrm{P}, \end{cases}$$

in which

$$\mathrm{M}'_{ij}=|\Omega_g|^{-1}\int_{\Gamma g} y_j.m'^i_k.n_{gk}\,ds.$$

Due to the symmetry of $\langle\ ,\ \rangle_1$, $\mathbf{M}'$ is a symmetric real tensor which depends on the frequency through $(1/2)(\omega_r/\omega)^2$. It can again be shown that the diagonal terms $\mathrm{M}'_{ii}$ (no summation on $i$) satisfy the inquality $\mathrm{M}'_{ii}\geqq\beta/(1-\beta)$.

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